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M. Sweany, A. Bernstein, N. Bowden, S. Dazeley, R. Svoboda

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Special Nuclear Material Detection with a Water Cherenkov based Detector

Melinda Sweany, Adam Bernstein, Nathaniel Bowden, Steven Dazeley, and Robert Svoboda

Abstract—Fission events from Special Nuclear Material (SNM), such as highly enriched uranium or plutonium, produce a number of neutrons and high energy gamma-rays. Assuming the neutron multiplicity is approximately Poissonian with an average of 2 to 3, the observation of time correlations between these particles from a cargo container would constitute a robust signature of the presence of SNM inside. However, in order to be sensitive to the multiplicity, one would require a high total efficiency. There are two approaches to maximize the total efficiency; maximizing the detector efficiency or maximizing the detector solid angle coverage. The advanced detector group at LLNL is investigating one way to maximize the detector size. We are designing and building a water Cerenkov based gamma and neutron detector for the purpose of developing an efficient and cost effective way to deploy a large solid angle car wash style detector. We report on our progress in constructing a larger detector and also present preliminary results from our prototype detector that indicates detection of neutrons.

I. MOTIVATION

Legitimate cross border trade involves the transport of an enormous number of very large cargo containers. In order to verify that these containers are not transporting SNM without impeding legitimate trade, there is a need for fast, highly efficient, and large detectors that are relatively inexpensive. The detectors need to produce a consistent response to SNM so that their effectiveness is not reduced by false positive or negative detections. They also need to have limited sensitivity to other types of background radiation, such as general cosmic ray induced background or Naturally Occurring Radioactive Material (NORM) present in certain legitimate forms of cargo. Both of these may contribute to false signals or reduce sensitivity to real SNM.

SNM can either spontaneously fission or be induced to do so by an external source of gamma rays or neutrons. Since cargo containers are large, the can contain a significant amount of shielding. The only fission products likely to be penetrating enough to exit the container and interact with a detector are neutrons or high energy (> 3MeV) gammarays. We propose that a water Cherenkov detector doped with a neutron capturing agent (such as GdCl₃ salt) would be an ideal detector for this application. Such a detector has a number of advantages; it would be relatively inexpensive, non-flammable, environmentally safe and easy to operate. Water is also insensitive to fast neutrons produced by cosmic-ray muon interactions; in organic scintillator, fast neutrons are capable

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M. Sweany and R. Svoboda are also at the University of California, Davis S. Dazeley, A. Bernstein, and N. Bowden are at Lawrence Livermore National Laboratory

of producing a correlated signal via proton recoil, followed by neutron capture. Thermal neutron capture on Gadolinium has an extremely high cross section (49000 barns). On capture, a gamma-ray shower with energies adding to approximately 8 MeV is produced. The subsequent Cherenkov radiation is detectable by ordinary PMTs. Super-Kamiokande and SNO have shown that the Cherenkov process can generate enough photons in water to detect neutron captures or gamma-rays with an energy of approximately 3 MeV or greater, so long as the photocathode coverage is high (40%). With the addition of highly reflective white walls, it should be possible to detect gammas of a few MeV or more with photocathode coverage of only $\sim 10\%$.

II. PROTOTYPE DETECTOR

We have designed and built a small (250 liter) water Cherenkov based prototype detector and tested its ability to detect high energy fission gamma-rays and neutrons [1]. A schematic of this detector is shown in Fig. 1. The detector was tested using a ²⁵²Cf fission source. We present preliminary results from our prototype that indicates we have detected neutrons. This encouraging result has led to a project to build a larger water based detector, approximately 4 cubic meters in size. After measuring the efficiency of our detector, we will investigate some related issues that may affect the performance. One issue will be the effect of high energy gamma-rays from the local environment and neutrons due to the passage of muons either near or through our detector, which will become more significant as our detector becomes larger. We explore methods of increasing our signal over these backgrounds with a full Geant4 detector simulation.

The prototype detector consists of two separate acrylic tanks; a small tank sits on top of a larger tank. An O-ring between the two tanks seals the volume of the lower tank. This lower tank (1m x .5m x .5m) contains ultra pure, sterilized water doped with 0.2% GdCl₃. The lower tank forms the main 250-liter active target volume of the detector. It is fitted with a small expansion volume and airlock so that the target remains full (i.e. optically coupled to the top tank) and closed to outside air despite ambient air pressure variations. The upper tank contains ultra pure, sterilized and deionized water without the Gd dopant, and eight downward facing 8 inch ETL 9354kb PMTs. The ETL PMTs have a relatively high quantum efficiency at short wavelengths ($\sim 30\%$), which is particularly advantageous for Cherenkov light detection. The PMTs are individually shielded from magnetic field effects by 8 inch diameter cylinders of mu-metal. The PMT and target volumes

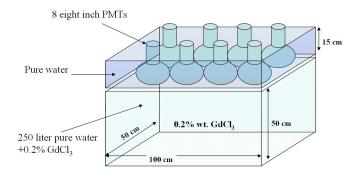


Fig. 1. Conceptual design of the prototype LLNL water Cherenkov based detector.

are separated to prevent exposure of the GdCl₃ doped water to the mu-metal surface; iron has been shown to react with GdCl₃ in water, reducing the water's clarity over time [2].

Several features are built into the design of the detector in order to maximize detection efficiency of Cherenkov photons. The tanks are constructed with UV transmitting acrylic; reflection was achieved by a combination of total internal reflection off the acrylic/air boundary and UV reflective 1073B Tyvek [3], [4]. The photo-cathode coverage is 10%. With the concentration of GdCl $_3$ at 0.2%, the large neutron capture cross section at thermal energies reduces the mean neutron capture time to about 30 μ s from the 200 μ s typical of pure water due to neutron capture on hydrogen.

III. PROTOTYPE DETECTOR RESULTS

The summed PMT response of all eight PMTs due to a $^{252}\mathrm{Cf}$ source (55 $\mu\mathrm{Ci}$ or 2.4 x 10^5 neutrons/s) placed approximately a meter from the detector is shown in Fig. 2. The red dashed curve shows the spectrum with the source present, the blue dotted curve is without the source present. The raw event rate increased from 700 Hz to 7 kHz due to the presence of the source alone. The black curve is the difference between the two. There is a clear response to the source, increasing the high energy (greater than 10 photo-electrons) component relative to background.

To determine whether this response is due to neutron capture, the inter-event time is plotted to determine if there is a correlated signal. Fig 3 shows the inter-event time with no source (bottom curve) and with the 252Cf source (top curve). The bottom curve shows a flat exponential fit for uncorrelated inter-event times greater than $100\mu s$, representing the background trigger rate. There is a correlated signal in the 0-100 μ s range; this is due to correlated events in the background, such as neutrons and gammas produced in radiative muon capture or spallation events. In the top curve, the random trigger rate from inter-event times greater than 100 μ s is again fit to an exponential and subtracted from the correlated rate. The residuals, shown in Fig 4, are then fit with an exponential, giving a 28 μ s capture time. This agrees well with the expected capture time given a concentration of 0.1 % gadolinium [5], [6]. These results indicate a clear signal due to the source, consistent with neutron capture.

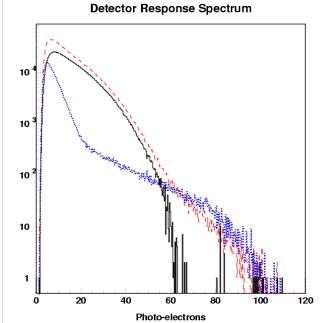


Fig. 2. Summed PMT response of prototype detector. Red dashed is with source present, blue dotted is without source present, and black is the difference between the two.

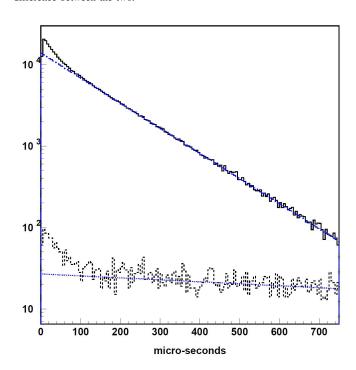


Fig. 3. The trigger rate with no source (bottom curve) and with a $^{252}\mathrm{Cf}$ source (top curve).

IV. FULL SCALE DETECTOR

We are currently building a larger detector, consisting of an approximately 4000 liter cylindrical tank of gadoliniumdoped water and two 20-PMT arrays at the bottom and top of the tank pointing inward. Several of the efficiency enhancing techniques from the prototype are included in the design, and new design elements for increasing the signal over background

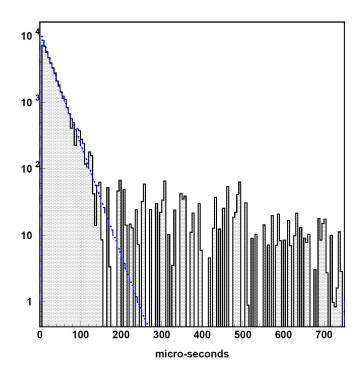


Fig. 4. Residuals of the correlated rate subtracted by the random trigger rate

are employed. One way to defeat backgrounds may be to group many small detectors together; the success of the prototype detector demonstrates that smaller detectors will work.

The larger detector is a cylindrical tank with a ~ 1.5 meter height and slightly less than 2 meter diameter. The tank holds roughly 3900 liters of water, and its inner surface will be lined with Teflon. There are 40 ten inch R7081 Hamamatsu PMTs, with individual mounts constructed out of acrylic, and housed in an acrylic frame. The PMTs are arranged in a circular pattern on the top and bottom of the tank, pointing inward. The quadrants of the detector will be divided by removable reflective sheets, connecting the top and bottom PMT arrays. The concentration of GdCl₃ will be between 0.1% and 0.5 %. The concept design of the larger detector is shown in Fig. 5.

A full simulation, including all major detector elements, has been developed in Geant4. Using this simulation, the increase in efficiency due to the wall reflectivity is demonstrated. The reflectivity of the Tyvek lining the inner surface of the tank was set to 90%, 50% and 10%. Neutrons were isotropically emitted from the center of the tank to determine the reflectivity response due to capture events in the detector. Fig. 6 shows the simulated response for these three reflectivities. The average number of PMT hits in the detector increase from 25 at 10% reflectivity to 40 at 90% reflectivity. We expect our detector's walls to have a reflectivity close to 90%.

Our simulation indicates that there is some difference in the light distribution of neutron capture events and background gamma events. The symmetry of the PMT signal is represented by the symmetric light distribution parameter (SLDP), given by

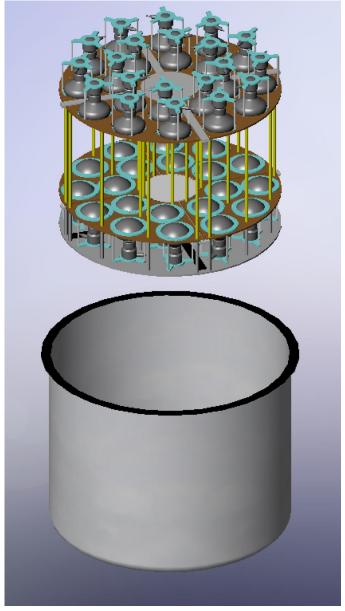


Fig. 5. Conceptual design of the prototype LLNL water Cherenkov based detector.

$$\sqrt{\frac{\sum_{i=0}^{N} x_i^2}{\left(\sum_{i=0}^{N} x_i\right)^2} - \frac{1}{N}}$$
 (1)

where N is the number of PMTs, and x_i is the number of photo-electrons in the ith PMT. This parameter is a measure of the standard deviation of PMT signals, weighted so that it is between 0 and 1. If all PMTs have the same number of photo-electrons, the parameter gives 0; if the entire signal is in only one PMT, the parameter will approach 1.

Using the detector simulation to determine the discrimination power of the SLDP, neutrons isotropically emitted from a point a meter to the right of the detector's center, or 6

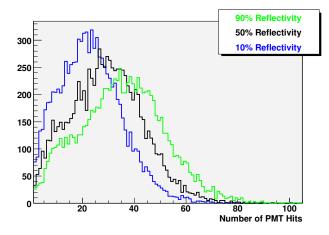


Fig. 6. The simulated detector response for various wall reflectivities.

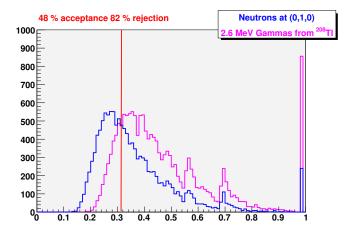


Fig. 7. SLDP for neutrons (blue) and background gammas (pink).

cm from the edge, are compared to 2.6 MeV gammas with random positions and momentums from outside the detector. In Fig. 7, the blue curve is the SLDP for neutrons and the pink curve is the SLDP for background gammas. Certain features of the distribution are more visible for the background gammas than for the neutrons, there are discernable peaks at 0.56 and 0.7, due to the signal being in only three and two PMTs respectively, as well as a large amount of events at nearly 1, corresponding to events where the signal is in only one PMT. The background distribution is also shifted towards one with respect to the neutron distribution, indicating less symmetry in the PMT signals than for neutron events. Cutting events with a SLDP of greater than 0.31 in the simulation will accept 48% of the signal neutrons, while rejecting 82% of the background gammas. This may become a more powerful discrimination with smaller detectors grouped together.

Using methods to increase efficiency in the prototype detector along with some new ones, in addition to some new ways to discriminate signal events from background, we hope to demonstrate detection of correlated neutron events from fission events with a larger water Cherenkov detector. Detector assembly is on schedule to begin in January 2009.

V. ACKNOWLEDGMENTS

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REFERENCES

- [1] arXiv:0808.0219v1
- [2] W. Coleman, A. Bernstein, S. Dazeley and R. Svoboda, Submitted to Nuclear Instruments and Methods in Physics Research A
- [3] Directional Reflectance Measurements (DR) on Two special sample Materials-Final Report, SOC-R950-001-0195, Prepared for the University of California, Irvine, School of Physical Sciences Irvine, California 92717-4675, January 1995.
- [4] Directional Reflectance (DR) Measurements on Five (5) UCI Supplied Sample Materials, SOC-R1059-001-0396, Prepared for the University of California, Irvine, School of Physical Sciences Irvine, California, March 1996
- [5] M. Apollonio et. al., European Phys. J. C27 331-374 (2003)
- [6] I A. G. Piepke, S. W. Moser, and V.M. Novikov, Nucl. Instr. and Meth. A 432 392-398 (1999)